

Temperature for a dynamic spin ensemble

Pui-Wai Ma,^{1,2} S. L. Dudarev,² A. A. Semenov,^{1,*} and C. H. Woo^{1,†}

¹*Department of Electronic and Information Engineering, The Hong Kong Polytechnic University, Hong Kong SAR, China*

²*EURATOM/CCFE Fusion Association, United Kingdom Atomic Energy Authority, Abingdon, Oxfordshire OX14 3DB, United Kingdom*

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In molecular dynamics simulations, temperature is evaluated, via the equipartition principle, by computing the mean kinetic energy of atoms. There is no similar recipe yet for evaluating temperature of a dynamic system of interacting spins. By solving semiclassical Langevin spin-dynamics equations, and applying the fluctuation-dissipation theorem, we derive an equation for the temperature of a spin ensemble, expressed in terms of dynamic spin variables. The fact that definitions for the kinetic and spin temperatures are fully consistent is illustrated using large-scale spin dynamics and spin-lattice dynamics simulations.

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I. INTRODUCTION

Temperature is the most fundamental quantity in statistical mechanics. In macroscopic equilibrium thermodynamics it is evaluated by differentiating entropy with respect to the energy of the system [1]. In experiment and microscopic molecular dynamics simulations it is probed by monitoring the mean kinetic energy of atoms [2–5] via

$$T = \frac{2}{3k_B N} \sum_i \frac{\langle \mathbf{p}_i^2(t) \rangle_t}{2m}. \quad (1)$$

Equation (1) refers to temperature associated with the excitation of translational degrees of freedom of particles described by an arbitrary Hamiltonian, where the kinetic energy E_K is a quadratic function of the generalized momenta $\mathbf{p}_i(t)$ [6]. For a classical Hamiltonian ensemble of interacting particles, temperature can also be evaluated by computing the configuration averages of forces acting between the particles [7–9]. The fact that Eq. (1) does not require differentiating the Hamiltonian makes it the most convenient and most often used recipe for monitoring temperature, through which the notion of temperature can be generalized even to nonequilibrium configurations, like the turbulent flow of liquids [10] or high-energy collision cascades [11].

Generalizing Eq. (1) to the case of a dynamic spin system is complicated by the fact that a generic spin Hamiltonian, such as the Heisenberg Hamiltonian $H = -\frac{1}{2} \sum_{i,j} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j$, does not have the form to which the existing methods [6–9] can be readily applied. Having a recipe for monitoring the spin temperature is necessary for analyzing a large variety of dynamic relaxation modes involving interactions between the spin, charge, and lattice degrees of freedom observed in experiments [12–17] and simulations [18–22]. To understand these interactions, and to study the corresponding relaxation modes, it would be useful to have a method, conceptually similar to Eq. (1), for evaluating the thermodynamic param-

eters of a spin ensemble, such as its temperature, directly from the variables characterizing its dynamic state. No such method presently exists.

A canonical spin system can be brought into equilibrium with a thermostat using the Langevin stochastic exchange field algorithm [23], subject to conditions imposed by the fluctuation-dissipation theorem (FDT) [24–27]. For example, in a spin-lattice dynamics simulation [21], interaction with a thermostat, described by some suitably chosen Langevin stochastic forces, drives spin orientations asymptotically to the equilibrium Gibbs distribution. The inverse problem, namely, of how to evaluate the spin temperature from the known orientations of all spins, is still awaiting solution.

In this paper, we derive an equation for the temperature of a spin ensemble expressed in terms of its dynamic state variables. The derivation involves investigating equilibrium solutions of semiclassical Langevin dynamic equations, and applying the FDT. The internal consistency between the formulas for spin temperature derived in this study, and for the *kinetic* temperature [Eq. (1)] referring to the excitation of translational degrees of freedom, is illustrated using large-scale spin and spin-lattice dynamics simulations.

II. LANGEVIN EQUATIONS AND THE SPIN TEMPERATURE

A. Particle case

Before addressing the spin case, we first investigate the well-known limit where particles, not interacting between themselves, move under the action of random dissipative forces. The Langevin equations for freely moving atoms are [24–27],

$$\frac{d\mathbf{p}_i}{dt} = -\frac{\gamma_l}{m} \mathbf{p}_i + \mathbf{f}_i(t), \quad (2)$$

where γ_l is the coefficient of dissipative drag for the translational, or lattice, degrees of freedom, and $\mathbf{f}_i(t)$ are the delta-correlated random forces satisfying conditions $\langle \mathbf{f}_i(t) \rangle = 0$ and $\langle f_{i\alpha}(t) f_{j\beta}(t') \rangle = \mu_l \delta(t-t') \delta_{ij} \delta_{\alpha\beta}$, where Greek symbols α and β denote the Cartesian components of a vector.

*Permanent address: Institute for Nuclear Research, Russian Academy of Sciences, 60-th October Anniversary Prospect 7a, 117312 Moscow, Russian Federation.

†Corresponding author: chung.woo@polyu.edu.hk

From the Langevin Eqs. (2) it follows that

$$\frac{d\langle E_K \rangle}{dt} = \frac{d}{dt} \sum_i \frac{\langle \mathbf{p}_i^2 \rangle}{2m} = - \sum_i \frac{\gamma_i \langle \mathbf{p}_i^2 \rangle}{m^2} + \sum_i \frac{1}{m} \langle \mathbf{p}_i(t) \cdot \mathbf{f}_i(t) \rangle. \quad (3)$$

Here E_K is the total kinetic energy of the system. The ensemble average is taken over all possible realizations of the stochastic forces. Substituting the formal solution of Eq. (2) into $\langle \mathbf{p}_i(t) \cdot \mathbf{f}_i(t) \rangle$, we find

$$\begin{aligned} \langle \mathbf{p}_i(t) \cdot \mathbf{f}_i(t) \rangle &= \frac{\gamma_i}{m} \int_0^t \langle \mathbf{p}_i(t') \cdot \mathbf{f}_i(t') \rangle dt' + \int_0^t \langle \mathbf{f}_i(t') \cdot \mathbf{f}_i(t') \rangle dt' \\ &= \frac{3\mu_l}{2}, \end{aligned} \quad (4)$$

The first integral in the middle of Eq. (4) vanishes because the argument of the integral $\langle \mathbf{p}_i(t') \cdot \mathbf{f}_i(t') \rangle$ is equal to zero everywhere in the interval $t' \in [0, t)$, except for the point $t' = t$ where it has a finite magnitude, unlike the second term, which integrates to give $3\mu_l/2$. Substituting Eq. (4) into the last term of Eq. (3) gives

$$\frac{d\langle E_K \rangle}{dt} = - \frac{2\gamma_l}{m} \sum_i \left(\frac{\langle \mathbf{p}_i^2 \rangle}{2m} - \frac{3\mu_l}{4\gamma_l} \right). \quad (5)$$

At equilibrium, the system is stationary and the time derivative of the mean kinetic energy in the left-hand side of the equation must vanish. In this limit, the right-hand side of Eq. (5) gives

$$\frac{1}{N} \sum_i \frac{\langle \mathbf{p}_i^2 \rangle}{2m} = \frac{3\mu_l}{4\gamma_l} = \frac{3}{2} k_B T, \quad (6)$$

where we equated the ensemble and the time average values, and evaluated the mean kinetic energy using the Gibbs distribution. The fact that μ_l and γ_l are related via $\mu_l = 2\gamma_l k_B T$ constitutes the FDT for the Langevin particles [24–27]. We note that Eq. (6) has the same form as Eq. (1), in agreement with the equipartition principle.

Equation (1) remains valid for the case of interacting particles. A Hamiltonian for interacting particles is

$$H = \sum_i \frac{\mathbf{p}_i^2}{2m} + U(\mathbf{R}), \quad (7)$$

where $U(\mathbf{R})$ is the potential energy of interaction between the atoms, and $\mathbf{R} = \{\mathbf{R}_i\}$ are the coordinates of atoms. The corresponding Langevin equations of motion are

$$\begin{aligned} \frac{d\mathbf{p}_i}{dt} &= - \frac{\partial U}{\partial \mathbf{R}_i} - \frac{\gamma_l}{m} \mathbf{p}_i + \mathbf{f}_i \\ \frac{d\mathbf{R}_i}{dt} &= \frac{\mathbf{p}_i}{m}. \end{aligned} \quad (8)$$

The change of the *total* energy with respect to time is

$$\begin{aligned} \frac{dE}{dt} &= \sum_i \left(\frac{\mathbf{p}_i \cdot d\mathbf{p}_i}{m \, dt} + \frac{\partial U}{\partial \mathbf{R}_i} \frac{d\mathbf{R}_i}{dt} \right) \\ &= \sum_i \left[\frac{\mathbf{p}_i}{m} \left(- \frac{\partial U}{\partial \mathbf{R}_i} - \frac{\gamma_l}{m} \mathbf{p}_i + \mathbf{f}_i \right) + \frac{\partial U}{\partial \mathbf{R}_i} \frac{\mathbf{p}_i}{m} \right] \\ &= \sum_i \left(- \frac{\gamma_l \mathbf{p}_i^2}{m^2} + \frac{\mathbf{p}_i \cdot \mathbf{f}_i}{m} \right). \end{aligned} \quad (9)$$

Comparing Eqs. (9) and (3), we see that the right-hand sides of both equations are identical. Using the same argument as for the case of non-interacting particles, we arrive at the relation between the momenta and kinetic temperature given by Eq. (1).

B. Spin case

Adopting an approach similar to that outlined above, we consider a generic Heisenberg Hamiltonian describing a broad class of spin systems [28],

$$H = - \frac{1}{2} \sum_{i,j} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j - \mathbf{H}_{ext} \cdot \sum_i \mathbf{S}_i, \quad (10)$$

where $\mathbf{H}_{ext} = -g\mu_B \tilde{\mathbf{H}}_{ext}$ is the effective external field, $\tilde{\mathbf{H}}_{ext}$ is the external magnetic field, g is the electronic g factor, μ_B is Bohr magneton, \mathbf{S}_i is a spin vector, and J_{ij} is the exchange interaction parameter involving spins i and j . J_{ij} is a function of the geometry of the lattice and the electronic structure of the material [28]. The Hamiltonian equations of motion for dynamic variables \mathbf{S}_i can be derived using the Poisson brackets method [29], namely,

$$\frac{d\mathbf{S}_i}{dt} = \frac{i}{\hbar} [H, \mathbf{S}_i] = \frac{1}{\hbar} \mathbf{S}_i \times \left(\sum_k J_{ik} \mathbf{S}_k + \mathbf{H}_{ext} \right) \equiv \frac{1}{\hbar} \mathbf{S}_i \times \mathbf{H}_i. \quad (11)$$

Here the effective exchange field acting on spin i is $\mathbf{H}_i \equiv \sum_k J_{ik} \mathbf{S}_k + \mathbf{H}_{ext}$. This effective field includes both the internal and external fields. We note that Eq. (11) retains the same form for both the interacting, or non-interacting, spin system.

In the presence of a random exchange field $\mathbf{h}_i(t)$ the spin system becomes nonconservative. The Langevin equations of motion for spins take the form [21]

$$\frac{d\mathbf{S}_i}{dt} = \frac{1}{\hbar} [\mathbf{S}_i \times (\mathbf{H}_i + \mathbf{h}_i(t)) - \gamma_s \mathbf{S}_i \times (\mathbf{S}_i \times \mathbf{H}_i)], \quad (12)$$

where γ_s is a dimensionless damping constant, which is introduced to compensate the effect of random noise $\mathbf{h}_i(t)$ [21]. The noise term $\mathbf{h}_i(t)$ in Eq. (12) is assumed to be delta correlated, satisfying the conditions $\langle \mathbf{h}_i(t) \rangle = 0$ and $\langle h_{i\alpha}(t) h_{j\beta}(t') \rangle = \mu_s \delta_{ij} \delta_{\alpha\beta} \delta(t - t')$. Here μ_s is a parameter characterizing the amplitude of the random noise, and the Greek symbols α and β again denote the Cartesian components of a vector. Equation (12) is similar to that proposed by Brown [23], who used a slightly different dissipation term $-\gamma_s \hbar \mathbf{S}_i \times d\mathbf{S}_i/dt$. The fact that to a first approximation $d\mathbf{S}_i/dt$ equals $(\mathbf{S}_i \times \mathbf{H}_i)/\hbar$ and that in Eq. (12) the right-hand side does not contain the time derivative of dynamic spin variables, makes

it consistent with the Langevin treatment of motion of particles described by Eq. (8).

The rate of variation of the ensemble-averaged total energy of the spin system, according to Eq. (10) and (12), is

$$\begin{aligned} \left\langle -\frac{1}{2} \sum_{i,j} J_{ij} \frac{d(\mathbf{S}_i \cdot \mathbf{S}_j)}{dt} \right\rangle &= - \sum_i \left\langle \mathbf{H}_i \cdot \frac{d\mathbf{S}_i}{dt} \right\rangle \\ &= -\frac{1}{\hbar} \sum_i \left[\langle \mathbf{H}_i \cdot (\mathbf{S}_i \times \mathbf{h}_i(t)) \rangle \right. \\ &\quad \left. + \gamma_s \langle |\mathbf{S}_i \times \mathbf{H}_i|^2 \rangle \right]. \end{aligned} \quad (13)$$

Here the ensemble average is taken over all possible realizations of the stochastic fields $\mathbf{h}_i(t)$. Evaluating the averages according to [30,31], we find

$$\langle \mathbf{H}_i \cdot (\mathbf{S}_i \times \mathbf{h}_i(t)) \rangle = -\frac{\mu_s}{\hbar} \langle \mathbf{S}_i \cdot \mathbf{H}_i \rangle, \quad (14)$$

Substituting this into Eq. (13), we arrive at

$$\frac{d\langle E \rangle}{dt} = \frac{1}{\hbar} \sum_i \left[\frac{\mu_s}{\hbar} \langle \mathbf{S}_i \cdot \mathbf{H}_i \rangle - \gamma_s \langle |\mathbf{S}_i \times \mathbf{H}_i|^2 \rangle \right]. \quad (15)$$

At thermal equilibrium, the system is stationary and the left-hand side of Eq. (15) must vanish. Since μ_s and γ_s are related by the FDT condition $\mu_s = 2\gamma_s \hbar k_B T$ (see Ref. [23] and Appendix), Eq. (15) leads to the following equation for the spin temperature, expressed in terms of dynamic spin variables $\mathbf{S}_i(t)$:

$$T = \frac{\mu_s}{2\gamma_s \hbar k_B} = \frac{\left\langle \sum_i |\mathbf{S}_i \times \mathbf{H}_i|^2 \right\rangle}{2k_B \left\langle \sum_i \mathbf{S}_i \cdot \mathbf{H}_i \right\rangle} = \frac{\left\langle \sum_i |\mathbf{S}_i(t) \times \mathbf{H}_i(t)|^2 \right\rangle_t}{2k_B \left\langle \sum_i \mathbf{S}_i(t) \cdot \mathbf{H}_i(t) \right\rangle_t}. \quad (16)$$

Equation (16) is the central result of this paper. In deriving this equation we used the fact that averaging over all the possible realizations of the stochastic field $\mathbf{h}_i(t)$ is equivalent to averaging over the statistical ensemble. Since J_{ij} are parameters characterizing a given material, the temperature T of a spin system in Eq. (16) is completely determined by the statistical distribution of the spins, in exactly the same way as the temperature of a system of moving particles is determined by its dynamics in Eq. (1). A striking difference between Eq. (16) and (1), however, is that the spin temperature defined by Eq. (16) can in principle be positive or *negative*, in agreement with Ref. [32] (the latter case is realized, for example, if the orientations of spins \mathbf{S}_i are opposite to those of the effective fields \mathbf{H}_i), whereas the kinetic temperature Eq. (1) for an atomic ensemble is positive definite. Of course, in equilibrium the temperature of a spin system is equal to that of the lattice, and has to be positive.

We also note that since the concept of temperature is thermodynamic, one needs to exercise caution when applying it to a non-equilibrium configuration. The issue of how to define temperature for a nonequilibrium situation is controversial [9] and is beyond the scope of the present paper. Nevertheless, under conditions that make local equilibrium a valid

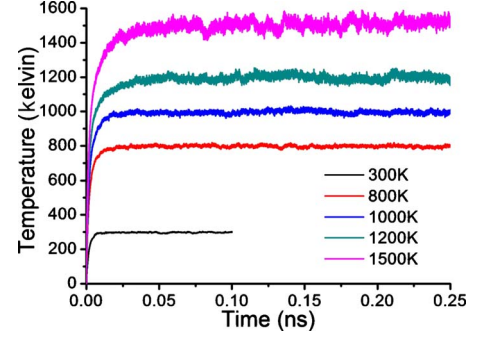


FIG. 1. (Color online) Evolution of temperature for a dynamic spin system interacting with a Langevin thermostat. Asymptotically, the spin temperature given by Eq. (16) approaches the temperature of the thermostat.

assumption, such as in the case where the global rate of energy change is much slower than that of the local equilibration processes, one may still assign the notion of temperature to a local dynamic set of variables according to Eq. (16). In such cases Eq. (15) shows that the empirical temperature assigned in this way still satisfies the fluctuation-dissipation condition, as required for a system in thermal equilibrium in that instance.

III. THREE CASE STUDIES

In what follows, we consider three examples where the spin temperature, evaluated using Eq. (16), can be compared with what is expected in terms of evolution and relaxation of the system. In the first case study, spin-dynamics (SD) simulations are performed for bcc iron, using the exchange coupling parameters J_{ij} chosen for ferromagnetic BCC iron according to Ref. [21]. We consider a spin ensemble realized on a $20a \times 20a \times 20a$ ($a = 2.8665$ Å) bcc lattice containing 16 000 atomic spins [21,33], with periodic boundary conditions applied in the x , y , and z directions. The initially collinearly ordered spin system is heated up and maintained at various preset temperatures by a Langevin thermostat [21]. No external field is applied. The dynamic temperatures of the spin systems evaluated using Eq. (16) are plotted as functions of time in Fig. 1. In each simulation, the spin temperature defined by Eq. (16) can be seen to rise from 0 K to an asymptotic value equal to the temperature of the canonical spin system as preset by the thermostat. The temperature fluctuations seen in Fig. 1 are due to limitations associated with the finite simulation cell size. Interestingly, this example suggests that if the temperature transient is sufficiently slow, Eq. (16) may also be used to monitor the thermal equilibration process of the spin system similarly to Eq. (1), the use of which in molecular dynamics simulations for this purpose is well established.

In the second case study, we perform spin-lattice dynamics (SLD) simulations [21,33] for a spin-lattice system with the rigid-lattice constraint removed. The temperature of the spin subsystem is controlled with a Langevin thermostat, but the temperature of the lattice is left unconstrained, with parameter γ_l set to zero. The lattice and spin subsystems are

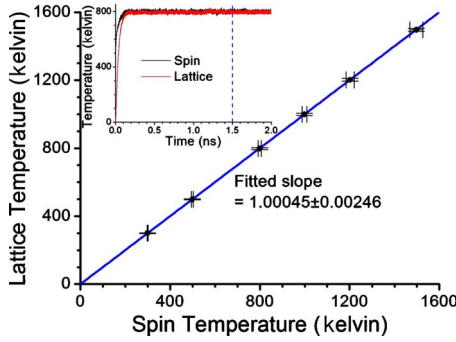


FIG. 2. (Color online) Lattice temperature [Eq. (1)] plotted against spin temperature [Eq. (16)] at equilibrium for a series of spin-lattice dynamics simulations. The Langevin thermostat is applied only to the spin subsystem. No temperature control applied to the lattice subsystem. Inset: the corresponding spin-lattice relaxation dynamics.

dynamically coupled via the coordinate-dependent exchange coupling function $J_{ij}(\mathbf{R})$ in the Heisenberg Hamiltonian Eq. (10). We anticipate that at equilibrium the spin temperature, evaluated using Eq. (16), and the lattice temperature, evaluated using Eq. (1), must be equal. Using the same 16 000 atoms simulation cell as before, and applying the stress-free boundary conditions, we equilibrate the spin-lattice system. The inset in Fig. 2 shows the results of “temperature measurements” involving Eq. (1) and (16) for the lattice and spin systems performed during the thermalization process. These “measured” temperatures can be seen to separately approach the temperature of the spin thermostat. At $t=1.5$ ns, we switch off the spin thermostat, turning the spin-lattice system into a microcanonical, i.e., an NVE, ensemble. The temperatures measured by Eq. (1) and (16) can be seen to remain steady, fluctuating about their average values. We note that these fluctuations are not related to the coupling between the lattice and the spin subsystems, but are intrinsic due to the large multiplicity of states of the spin system which have the same total energy but different spin orientations. In this regard, basic thermodynamics dictates that the temperature is determined by the change in the number of such states, i.e., the entropy, as a function of the energy of the system. We note that this is true for the fluctuations of both the spin and the lattice system; no matter whether they are coupled or decoupled. Figure 2 shows the average temperatures and their standard deviations for the lattice and the spin subsystems according to Eq. (1) and (16). The 45° straight line on which the data points lie shows very well that these two temperatures are equal at equilibrium and that Eq. (1) and (16) are consistent with each other.

Finally, we consider the microcanonical relaxation of an initially spatially and thermally heterogeneous spin system. The initial condition for the simulation illustrated in Fig. 3 is set by bringing into contact the two spin subsystems equilibrated at 300 and 800 K, respectively, occupying the left and right halves of the simulation cell, subject to periodic boundary conditions. Experimentally, a nonequilibrium spin configuration similar to that shown in Fig. 3 can be generated using a laser or a microwave pulse. The evolution of the spin system is followed using a microcanonical spin dynamics

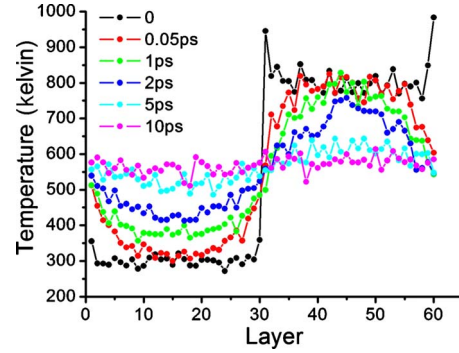


FIG. 3. (Color online) Relaxation dynamics for a spin system with an initial temperature gradient. Evolution of the system is followed using microcanonical spin dynamics simulations. The local spin temperature [Eq. (16)] is plotted as a function of the distance to the interface between the two initially separate parts of the system. The profiles reach equilibrium on the time scale of ~ 10 ps.

simulation. Since the total energy is conserved, the equilibration of the spin system takes place through the energy and angular momentum exchange between the neighboring volumes. In the simulation, the local energy, temperature [defined according to Eq. (16)], and magnetization are evaluated for each atomic layer parallel to the interface between the two subsystems. The profiles of the local temperatures are shown in Fig. 3. All the three quantities follow similar trends, approaching the equilibrium and becoming spatially homogeneous at the temperature of ~ 550 K on the time scale of approximately 10 ps. This simulation shows that the spin temperature, defined by Eq. (16), provides a useful means for monitoring the relaxation processes in the spin subsystem, similar to the way in which the notion of the local lattice temperature Eq. (1) is applied to understanding the microscopic dynamics of turbulence [10] or high-energy collision cascades [11].

IV. SUMMARY AND CONCLUSIONS

So far, there has been no practical recipe available for relating the temperature and the dynamic state of an ensemble of interacting spins. This issue is becoming increasingly important, as large scale simulations of spin ensembles evolve into tools for predictive exploration of magnetic phenomena on the nanoscale. This paper offers a closed-form expression that can be used for evaluating the thermodynamic temperature for a system of interacting spins in terms of the ensemble means of combinations of spin vectors. This expression was derived by solving the semi-classical Langevin-type equations of motion at equilibrium for the spins, and by using the fluctuation-dissipation theorem. The internal consistency between our spin-temperature expression [Eq. (16)] and the kinetic lattice temperature expression [Eq. (1)] is proven for the three case studies involving spin and spin-lattice dynamics simulations. In the first case, spin dynamics simulations of thermalization processes show that the spin temperature calculated according to Eq. (16) asymptotically approaches that of the thermostat, in agreement with expectations. In the second example, the spin and the lattice

degrees of freedom are allowed to interact, and direct spin-lattice dynamics simulations show that spin temperature, defined in terms of spin dynamic variables, is fully consistent with the lattice temperature defined in terms of kinematic momenta. In the third case study, we investigate the microcanonical relaxation of an initially spatially and thermally heterogeneous spin system. The equilibrium temperature profile, which can only be obtained via Eq. (16) in this case, agrees with expectations. Through this analysis, we prove that the notion of dynamic spin temperature offers a useful insight into the microscopic dynamics of relaxation of an initially spatially heterogeneous nonequilibrium system.

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APPENDIX

The fluctuation and dissipation forces drive the spin ensemble asymptotically to equilibrium by changing the spin orientations. In principle, one could derive the FDT condition for the Langevin equation of motion for spins [Eq. (12)] following the procedure described by Brown [23]. Here, we provide a simple derivation that proves the fluctuation-dissipation relation (FDR). We investigate the case of a single spin interacting with stationary external field.

The Langevin equation of motion for a single spin in an external field, which resembles Eq. (12), has the form

$$\frac{d\mathbf{S}}{dt} = \frac{1}{\hbar} \{ \mathbf{S} \times [\mathbf{H}_{ext} + \mathbf{h}(t)] - \gamma_s \mathbf{S} \times (\mathbf{S} \times \mathbf{H}_{ext}) \}. \quad (\text{A1})$$

It can be rearranged into a sum of terms involving, or not involving, random noise, i.e., $\dot{\mathbf{S}} = \mathbf{F} + \underline{\mathbf{G}} \cdot \mathbf{h}$, where $\mathbf{F} = [\mathbf{S} \times \mathbf{H}_{ext} - \gamma_s \mathbf{S} \times (\mathbf{S} \times \mathbf{H}_{ext})] / \hbar$ and $\underline{\mathbf{G}}$ is a tensor with components $G_{\alpha\alpha} = 0$, $G_{\alpha\beta} = -S_\gamma / \hbar$, and $G_{\alpha\gamma} = S_\beta / \hbar$. The lower-case Greek indices α , β , and γ satisfy the cyclic permutation relations for x , y , and z .

We map the equations of motion to the Fokker-Planck equation [26,27],

$$\frac{\partial W}{\partial t} = - \sum_{\Theta} \frac{\partial}{\partial S_{\Theta}} (A_{\Theta} W) + \frac{1}{2} \sum_{\Theta, \Phi} \frac{\partial^2}{\partial S_{\Theta} \partial S_{\Phi}} (B_{\Theta\Phi} W), \quad (\text{A2})$$

where W is the energy distribution function, $A_{\alpha} = \lim_{\Delta t \rightarrow 0} \frac{1}{\Delta t} \langle \Delta S_{\alpha} \rangle$ and $B_{\alpha\beta} = \lim_{\Delta t \rightarrow 0} \frac{1}{\Delta t} \langle \Delta S_{\alpha} \Delta S_{\beta} \rangle$ are the drift

and diffusion coefficients, respectively. The upper-case Greek indexes Θ and Φ refer to the Cartesian components of a vector or a tensor.

According to Brown [23], the drift and diffusion coefficients can be expressed in terms of W_0 and $\underline{\mathbf{G}}$ as

$$\begin{aligned} A_{\alpha} &= F_{\alpha} + \frac{1}{2} \mu_s \sum_{\Theta, \Phi} \frac{\partial G_{\alpha\Phi}}{\partial S_{\Theta}} G_{\Theta\Phi} \\ B_{\alpha\beta} &= \mu_s \sum_{\Theta} G_{\alpha\Theta} G_{\beta\Theta}. \end{aligned} \quad (\text{A3})$$

After some algebra, we arrive at

$$\begin{aligned} \mathbf{A} &= \frac{1}{\hbar} [\mathbf{S} \times \mathbf{H}_{ext} - \gamma_s \mathbf{S} \times (\mathbf{S} \times \mathbf{H}_{ext})] - \frac{\mu_s}{\hbar^2} \mathbf{S} \\ B_{\alpha\beta} &= \mu_s (\delta_{\alpha\beta} |\mathbf{S}|^2 - S_{\alpha\beta}) / \hbar^2. \end{aligned} \quad (\text{A4})$$

It should be noted that not only γ_s , but also μ_s enter in the drift coefficient A_{α} . In the lattice case, only γ_l enters the expression for A_{α} and only μ_l enters $B_{\alpha\beta}$. Since random noise is delta correlated, and all the components of the linear momenta are independent, noise can in no way interact with other components of the momenta, and it does not enter the drift term. In the spin case, the effect of random noise goes into the drift term due to the presence of the cross product in the equations of motion, or equivalently the rotation nature of the spin motion. This allows the interaction of the noise at the current instant with the previous instant through other components of the spin vector.

In thermal equilibrium (i.e., $\partial W / \partial t = 0$), we identify the energy distribution for the spin system with the Gibbs distribution $W = W_0 \exp(-H / k_B T)$ where W_0 is a normalization constant and $H = -\mathbf{S} \cdot \mathbf{H}_{ext}$. Substituting the Gibbs distribution into the Fokker-Planck equation, we find

$$\frac{\partial W}{\partial t} = \frac{1}{\hbar} \left(\frac{\mu_s}{\hbar k_B T} - 2\gamma_s \right) \left(\frac{1}{2k_B T} |\mathbf{S} \times \mathbf{H}_{ext}|^2 - \mathbf{S} \cdot \mathbf{H}_{ext} \right) W. \quad (\text{A5})$$

Since the second bracket in the right-hand side contains variable quantities, the condition of thermal equilibrium implies that the right-hand side of Eq. (A5) vanishes if and only if

$$\mu_s = 2\gamma_s \hbar k_B T. \quad (\text{A6})$$

This means that if μ_s and γ_s are related by Eq. (A6), the Gibbs distribution solves the Fokker-Planck equation. This proves the FDR. The derivation being presented here can be generalized to the case of many-body interacting spins. In fact, the FDR should not change for an interacting spin system, provided that the way how the fluctuation and dissipation force enters the equations of motion remains the same.

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